

## **Project Summary of the NREL Amorphous Silicon Team**

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# Project Summary of the NREL Amorphous Silicon Team

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## ABSTRACT

The Amorphous Silicon Team at NREL has improved the properties of many materials, increased solar cell device performance, and improved the fundamental understanding of thin-film silicon based materials and devices since the last NCPV Program Review Meeting. In this paper we present a summary of the work of the team since that last meeting.

### 1. Introduction

Four main areas limit the large scale production of hydrogenated amorphous silicon (a-Si:H) as low-cost PV. These are:

- (a) A relatively low conversion efficiency (relative to other PV technologies)
- (b) The Staebler-Wronski Effect (material properties—and thus devices—degrade with illumination)
- (c) There is a lack of a suitable low bandgap material (a-Si:H is a wide bandgap material,  $\geq 1.7$  eV)
- (d) That the best a-Si:H based materials are grown at the lowest deposition rates (increasing production costs).

In spite of these limitations, a-Si:H modules are still produced at low cost by industry.

The efforts of the NREL Amorphous Silicon Team emphasize these four areas. However, it is not always straightforward to assign a particular effort to one particular category. For example, improvements in deposition techniques and understanding, as well as increased understanding of defects, can have implications to all for of the above areas.

### 2. Improving Device Efficiency

We grew a-Si:H, n-i-p solar cells on untextured stainless steel substrates. All layers were grown by the hot-wire chemical vapor deposition (HWCVD) technique, and the i-layers were  $\sim 1800$  Å thick. These cells have the best known open-circuit voltage ( $V_{oc} = 0.94$  V) for all-HWCVD cells, with a good fill factor (0.74) and a current density of 8-9 mA/cm<sup>2</sup>, as needed for a top cell if incorporated into a triple cell device. The improvement in  $V_{oc}$  ( $> 60$  mV higher than our previous work) was obtained by hydrogen-diluting the i-layer at the i-p interface. This produces materials that are close to the phase transition from amorphous to microcrystalline silicon. We also lowered the substrate temperature to 150°C for the i-layer (most likely widening the bandgap) and performed a brief atomic H-treatment to the i-layer before the p-layer growth [1].

Hydrogenated microcrystalline silicon ( $\mu$ c-Si:H) solar cells need thicker i-layers than a-Si:H cells due to a lower absorption coefficient ( $\geq 1$   $\mu$ m and  $\leq 0.4$   $\mu$ m, respectively). Therefore, HWCVD is promising for

growing  $\mu$ c-Si:H devices because good materials are grown at high deposition rates ( $R_d$ ) relative to other low-temperature ( $< 500^\circ\text{C}$ ) CVD technologies [2]. Using stainless steel substrates with Ag/ZnO back reflectors supplied by UniSolar, we have grown  $\mu$ c-Si:H solar cells with initial efficiencies ( $\eta_{int}$ ) as summarized in Table 1.

Table 1:  $\mu$ c-Si:H device summary

Ref. #	$R_d$ (Å/s)	$T_{fil}$ (°C)	$T_{sub}$ (°C)	$\eta_{int}$ (%)	$J_{sc}$ (mA/cm <sup>2</sup> )
[3]	10	1750	350	6.0	20
[4]	5	1800	300	6.5	22
[4]	8	1900	250	4.0	22

Although the lower filament temperatures produce—for the time being—superior device results, filament temperatures below 1900°C have severe alloying problems. This not only limits the useful life of a filament, but reproducibility becomes an issue. Therefore, we have recently started to focus our efforts on a deposition regime in which our filament lifetime and reproducibility are not at issue. By running our filaments at 13 amps each (1900°C), with a 7-minute hydrogen treatment between each device. In previous attempts to grow  $\mu$ c-Si:H devices with  $T_{fil} > 1900^\circ\text{C}$ , we could not fabricate functioning devices. In the current effort, we are fabricating devices with initial efficiencies of 4% and very good reproducibility. These results are obtained by going from 140 mTorr and 300°C for the i-layer growth, down to 60 mTorr and 250°C [4].

### 3. Increasing Stability

There is general acceptance that H is involved in the Staebler-Wronski Effect (SWE) in some capacity. In 1998, Howard Branz postulated a microscopic model that accounts quantitatively for the kinetics of defect creation and annealing due to light-induced emission and retrapping processes for mobile atomic H [5].

Recently, we have done measurements that show that the charge state of diffusing hydrogen in a-Si:H depends upon its position relative to the electronic Fermi level ( $E_f$ ). We grew i-n-i-p-i structures on crystalline silicon substrates with a thin, tracer layer of deuterium ( $2H > 10^{19}$  cm<sup>-1</sup>) at various positions between the n- and p-layers. The electric field (F) is above  $6 \times 10^4$  V/cm at each 2H layer. We then annealed the samples at various temperatures and times and observed asymmetries in the secondary ion mass spectrometry profiles of diffused 2H. The 2H diffusion is predominately toward the closest doped layer, indicating H is moving with F. Because the Si-H bond is neutral, the charged H state must be formed after it is emitted into the mobile state [6]. Recently, we have repeated these experiments on similar structures using deuterated silane

(rather than deuterium dilution of silane) and increased the 2H concentration to  $> 10^{21} \text{ cm}^{-3}$ . We are currently analyzing these results, but they show similar, albeit cleaner, results.

By exposing a-Si:H samples to UV illumination and then subsequently etching away a 700-to-1000-Å thick “damaged” layer, we observe an improvement in the photoconductivity stability against light-soaking. The treated samples begin with red-light photoconductivities inferior to that of a control sample that is only etched. After less than an hour of 1-sun red light-soaking, the photoconductivity of the etched-only control falls below that of the UV-etch treated sample. After 2 to 3 days light-soaking, the UV-etch films can have a photo-sensitivity 20% to 38% above their control. The UV-etch treatment also produces small improvements in the stabilized open-circuit voltage of Schottky barrier solar cells. We speculate that mobile hydrogen produced during UV illumination is penetrating the film and improving stability. However, we observe no corresponding improvement of defect optical absorption by constant photocurrent method spectroscopy [7, 8]. The kinetics solar cell degradation more closely match photoconductivity degradation than defect creation.

#### 4. Improving Narrow-Bandgap Materials

By decreasing our W filament diameter and both our substrate and filament temperatures, we have improved the quality of our narrow bandgap a-SiGe:H grown by HWCVD. We now grow a-SiGe:H with Tauc bandgaps below 1.5 eV, and a photoresponse equal to or better than our PECVD grown alloys. We enhanced the transport properties—as measured by the photoconductivity frequency mixing technique—relative to previous HWCVD results [9]. We reported the results of these measurements as well as small angle X-ray scattering (SAXS) and photoluminescence (PL) measurements at the 2<sup>nd</sup> International Conference on Cat-CVD (Hot-Wire CVD) Process in Denver, Colorado [10, 11]. We have found conditions that produce alloys with reduced microvoid density and improved H-bonding (strong Si-H and Ge-H and no Si-H<sub>2</sub> or Ge-H<sub>2</sub>). Yueqin Xu is presenting a paper at this conference detailing these improvements and will publish even more details at the upcoming Spring MRS meeting [12].

The alternate narrow bandgap material for thin Si solar cell applications is  $\mu\text{c-Si:H}$ . We have collaborated with various people to characterize the optoelectronic and structural properties of these materials. This includes X-ray diffraction (XRD), scanning electron microscopy (SEM), cross-sectional tunneling electron microscopy (TEM), atomic force microscopy (AFM), real-time spectroscopic ellipsometry (RTSE), Raman, room temperature conductivity, and PL. This is a complicated material that is not easily characterized, and simple conclusions are hard to define. This is due, in part, to the fact that the properties are sensitive to the growth conditions, substrate (template effects), and sample thickness. Because of space limitations in this publication, we provide the references to this work for

the interested reader. The effect of having a seed layer on highly  $\mu\text{c-Si}$  materials is reported in both references [2 and 13]. We have characterized the phase transition from amorphous to microcrystalline Si using RTSE. The degree of crystallinity of the film can be determined from the form of the dielectric function. We reported on two of these studies in references [14 and 15]. The methodology we developed in [15] is very useful for characterizing these materials. We also applied the combinatorial approach to this phase transition. Because films become more microcrystalline with increasing thickness (for fixed deposition conditions), we were able to grow a film “stripe” of varying thickness. Several stripes using different HWCVD growth conditions—such as hydrogen to silane dilution, chamber pressure, and substrate temperature—were used to grow multiple stripes on a single substrate. This material library of thickness-graded films has regions of amorphous, phase transition, and microcrystalline Si on a single stripe. Various ex-situ characterization were used to characterize these samples [16-18].

#### 5. Increasing Deposition Rates

We can grow “device quality” a-Si:H by HWCVD at deposition rates up to 130 Å/s, and have grown working solar cells from this material. The short-range order (from Raman), the medium-range order (from XRD), and the peak position of the H evolution peak do not vary with increasing deposition rate from 5 to  $>140$  Å/s (for films with similar H content). The structure is consistent with “device quality” a-Si:H deposited at low deposition rates. However, the degree of structural heterogeneity as measured by SAXS, increases by a factor of 100 over that for the best (compact) a-Si:H for deposition rates over 20 Å/s. This is most likely due to an increase in microvoids. The as-grown defect density increases with increasing deposition rate, however, the saturated defect density—after light-soaking—is invariant with deposition rate [19]. For films grown at deposition rates over 50 Å/s, there is a large red shift of the PL peak energy. The PL intensity is as high as that in typical low-deposition-rate films. It may be that highly strained bonds on void surfaces form broad conduction-band tail states that are responsible for the PL red shift [20].

When RTSE is used for in-situ characterization of the optical properties and surface roughness ( $R_s$ ) of HWCVD grown a-Si:H with deposition rates varying from 5 to 120 Å/s, we find that the early-time evolution of the  $R_s$  is remarkably similar for all deposition rates. During the first few Å of growth, there is a sharp increase in  $R_s$  as the a-Si:H nucleates in separate islands. This is followed by a reduction of  $R_s$  as these areas coalesce into a bulk film, which occurs at an average thickness of 100 Å. After coalescence, the  $R_s$  rises to a stable value that is dependent upon growth conditions with a general tendency for the  $R_s$  to increase with growth rate. However, neither the  $R_s$  nor the material electronic properties are unique for a given deposition rate. We observe a stronger correlation of film properties with  $R_s$  than with deposition rate; namely, a monotonic decrease in photo-response, and increase in optical gap, with increasing  $R_s$  [21].

## 6. Improving Deposition Techniques/Understanding

We continue to develop combinatorial techniques for the deposition and characterization of Si thin films. The application of a combinatorial approach to these materials areas can greatly accelerate the rate of discovery of new materials and growth regimes, as well as the testing of new device designs. In addition to the “combi” results reported above, we have published work on the combinatorial technique itself [22].

Since the last NCPV Program Review Meeting, we have incorporated RTSE into one of our HWCVD cross-reactors to study the optical properties of films as they grow. The instrument is a J.A. Woollam, Inc., M2000 visible and near IR rotating compensator ellipsometry using Woollam software for instrument control and data acquisition and analysis. We typically collect spectra from 255 to 1240 nm with an integration time of 200 to 500 ms during the nucleation phase of growth and 1 to 5 seconds during the later stages of growth, dependent upon film deposition rates. The angle of incidence is fixed by the chamber geometry at 70°. In addition to the RTSE results reported above, we have studied the growth of a-Si:H (no H<sub>2</sub> dilution) and found a distinct 100-Å-thick layer at the top of the growing film that we have termed the growth zone. This material “rides on top” of the growing film and has distinct optical properties from the bulk film below. It is interesting that optical properties of the growth zone are similar among various deposition conditions (e.g., substrate temperatures), whereas the optical properties of the bulk film left behind can be quite different [23].

## 7. Increasing the Understanding of Defects

We find experimental fingerprints of two distinct metastable defects created during illumination of a-Si:H. The well-studied, three fold-coordinated silicon dangling bond defect has an annealing activation energy ( $E_a$ ) of ~1.1-eV and dominates annealing experiments  $\geq 110^\circ\text{C}$ . The second defect created by illumination is the “primary recombination” (pr) center, which causes most of the light-induced photoconductivity decrease and dominates annealing experiments  $\leq 110^\circ\text{C}$ . Because the pr centers are created in linear proportion to the dangling bond defects, they are difficult to distinguish during degradation. In low-temperature annealing studies, we find that the pr center has an annealing activation energy of ~0.85 eV and is responsible for a sharp increase in photoconductivity (that is not seen in neutral dangling bond studies). There is also a surprising increase in the ~1.1-eV sub-gap optical absorption [24].

Measurements of capacitance transients due to charge-carrier emission from metastable defects in p-type a-Si:H p/n junction structures show that both holes and electrons can be metastably trapped in the p-layer. At 350 K, electrons and holes are emitted at the same rate, thereby defining the isokinetic temperature ( $T_{\text{iso}}$ ) for the Meyer-Neldel rule. The enthalpy changes for hole and electron emission are 0.94 and 0.51 eV, respectively. Charge emission rates are measured above and below

$T_{\text{iso}}$ . The entropy changes for hole and electron emission are 31 and 17 Boltzmann constants, respectively. Below  $T_{\text{iso}}$ , electrons are emitted faster than holes; above  $T_{\text{iso}}$  the reverse is true. These relative changes in emission rates are a direct consequence of the large entropy changes in the defect reactions [25, 26].

## 8. Conclusions

The NREL Amorphous Silicon Team continues to perform basic and applied science within the context of the international thin-silicon community. During the time since the last NCPV Program Review Meeting, the NREL team has collaborated with 21 individuals from nine different universities and two industries, from four countries in addition to the United States. There have also been collaborations with more than 15 people outside the team from the NCPV.

## 9. Acknowledgements

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